

Simulation of Upper Tropospheric CO₂ From Chemistry and Transport Models

Xun Jiang^{1*}, Qinbin Li¹, Mao-Chang Liang^{2,3,4}, Run-Lie Shia⁴, Moustafa T. Chahine¹,
Edward T. Olsen¹, Luke L. Chen¹, and Yuk L. Yung⁴

¹ Science Division, Jet Propulsion Laboratory, California Institute of Technology, USA

² Research Center for Environmental Changes, Academia Sinica, Taipei, Taiwan.

³ Graduate Institute of Astronomy, National Central University, Jhongli, Taiwan.

⁴ Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, USA.

* To whom all correspondence should be addressed. E-mail: xun@gps.caltech.edu

Submitted to GBC, Jun 30, 2007; Revised, Apr 2, 2008

Abstract:

The Caltech/JPL two-dimensional (2-D), three-dimensional (3-D) GEOS-Chem, and 3-D MOZART-2 chemistry and transport models (CTMs), driven respectively by NCEP2, GEOS-4 and NCEP1 reanalysis data, have been used to simulate upper tropospheric CO₂ from 2000 to 2004. Model results of CO₂ mixing ratios agree well with monthly mean aircraft observations at altitudes between 8 and 13 km [*Matsueda et al.*, 2002] in the tropics. The upper tropospheric CO₂ seasonal cycle phases are well captured by the CTMs. Model results have smaller seasonal cycle amplitudes in the southern hemisphere compared with those in the northern hemisphere, which are consistent with the aircraft data. Some discrepancies are evident between the model and aircraft data in the mid-latitudes, where models tend to underestimate the amplitude of CO₂ seasonal cycle. Comparison of the simulated vertical profiles of CO₂ between the different models reveals that the convection in the 3-D models is likely too weak in boreal winter and spring. Model sensitivity studies suggest that convection mass flux is important for the correct simulation of upper tropospheric CO₂.

1. Introduction

[1] The increasing level of atmospheric CO₂ has significant influence on the global climate changes [Dickinson and Cicerone, 1986]. It is very difficult to disentangle the contributions from different sources and sinks of atmospheric CO₂. Most inversions for the CO₂ sources and sinks are constrained by surface measurements [Fan *et al.*, 1998; Tans *et al.*, 1990; Suntharalingam *et al.*, 2003; Gurney *et al.*, 2004]. For example, the global three-dimensional (3-D) inverse modeling analysis of surface flask and oceanic CO₂ measurements by Tans *et al.* [1990] implied a significant carbon sink in the northern hemisphere (NH) terrestrial biosphere. The inversion of carbon fluxes shows sensitivity to CO₂ network configuration [Gloor *et al.*, 2000; Suntharalingam *et al.*, 2003]. In addition, the vertical transports in the models are also very important for the inversion. The Atmospheric Tracer Transport Model Intercomparison Project (TRANSCOM) was created to quantify the uncertainty in the atmospheric CO₂ inversions from atmospheric transport [Gurney *et al.*, 2003]. Some results from TRANSCOM and other models suggest that inversion results are also very sensitive to vertical transport in the tracer transport models [Law *et al.*, 1996; Fan *et al.*, 1998; Gurney *et al.*, 2004].

[2] Previous modeling studies [Randerson *et al.*, 1997; Kawa *et al.*, 2004] have primarily employed surface measurements of CO₂ in their analysis. They compared the seasonal cycle and trend of surface CO₂ with their model results. The upper tropospheric CO₂ concentrations, from *in situ* aircraft measurements, usually differ by ~5 ppmv relative to the surface concentrations [Anderson *et al.*, 1996; Nakazawa *et al.*, 1997]. Matsueda *et al.* [2002] have been measuring CO₂ mixing ratios biweekly since April 1993 aboard commercial airlines at 8-13 km altitudes over the western Pacific from Australia to Japan. This data set offers a unique opportunity to test the ability of chemistry and transport models (CTMs) in simulating the upper tropospheric CO₂. The retrievals of CO₂ mixing ratios from the Atmospheric Infrared Sounder (AIRS), with a high precision of ~1-2 ppmv [Chahine *et al.*, 2005; Chahine *et al.*, 2008], can provide the global map of the middle to upper tropospheric CO₂ on a weekly basis. There is significant spatiotemporal variability in AIRS CO₂, which is supported by the *in situ*

aircraft observations. It remains a challenge for the CTMs to simulate the spatiotemporal CO₂ variability in the middle to upper troposphere. AIRS CO₂ retrievals can be used for constraining the vertical transport in CTMs in the future [Chahine *et al.*, 2008].

[3] Using a two-dimensional (2-D) CTM, Shia *et al.* [2006] successfully simulated the seasonal cycle and trend of CO₂ in the upper troposphere. In this study, we will investigate instead how well global 3-D CTMs are able to simulate the seasonal cycle and trend of upper tropospheric CO₂. Surface emissions and vertical transport in CTMs are both very crucial for the correct simulation of CO₂. We will use two different boundary conditions to investigate the contribution of boundary conditions to the upper tropospheric CO₂. One is a boundary condition where the CO₂ surface mixing ratios are constructed with measurements from the GLOBALVIEW-CO₂ surface network. The other is with prescribed known CO₂ sources and sinks. To investigate the influence of vertical transport, we will compare results from GEOS-Chem and MOZART-2 with four different vertical transport schemes.

2. Models and Data

[4] The Caltech/JPL 2-D CTM [Shia *et al.*, 2006], 3-D GEOS-Chem [Suntharalingam *et al.*, 2004], and 3-D MOZART-2 [Horowitz *et al.*, 2003] are used to simulate CO₂. The 2-D CTM has 18 latitude zones, equally spaced from pole to pole. It has 40 vertical layers, equally spaced in log scale of pressure from the surface to the upper boundary at 0.01 hPa. Transport in the model is by the stream function and the horizontal and vertical diffusivities taken from Jiang *et al.* [2004]. The stream function is derived from the National Center for Climate Prediction (NCEP) Reanalysis 2 data [Jiang *et al.*, 2004]. For altitudes above 40 km where no NCEP data are available, we adopt the climatologically averaged circulation derived by Fleming *et al.* [2002]. There is a gradual merging of the two data sets between 30 and 40 km. An important feature of the 2-D CTM is its ability to reproduce the age of air in the stratosphere [Morgan *et al.*, 2004].

[5] GEOS-Chem (v7.3.3) is driven by the Goddard Earth Observing System (GEOS-4)

assimilated meteorological data from the NASA Global Modeling Assimilation Office (GMAO). For computation efficiency, we regridded the GEOS-4 data into 2° (latitude) \times 2.5° (longitude) in horizontal and 30 levels in vertical. It extends from the surface to about 0.01 hPa (~ 70 km). Advection is computed every 15 minutes with a flux-form semi-Lagrangian method [Lin and Rood, 1996]. Moist convection is computed using the GEOS convective, entrainment, and detrainment mass fluxes described by Allen *et al.* [1996a, 1996b]. The physics in the GEOS-4 analysis system are adopted from the National Center for Atmospheric Research (NCAR) Community Climate Model, Version 3 (CCM3) and Whole Atmosphere Community Climate Model (WACCM) with important modifications to make it suitable for data assimilation [Bloom *et al.*, 2005]. The deep convection scheme is based on Zhang and McFarlane [1995]. The shallow convection treatment follows Hack [1994]. The planetary boundary layer turbulence parameterization is from Holtslag and Boville [1993]. To investigate the influence of different vertical mixings on the upper tropospheric CO₂, we also force the GEOS-Chem model with the GEOS-3 reanalysis data, which employs the Relaxed Arakawa Schubert convection parameterization [Moorthi and Suarez, 1992].

[6] MOZART-2 is driven by the meteorological inputs every 6 hours from the NCEP Reanalysis 1 [Kalnay *et al.*, 1996]. Advection is computed every 20 minutes with a flux-form semi-Lagrangian method [Lin and Rood, 1996]. The horizontal resolution is 2.8° (latitude) \times 2.8° (longitude) with 28 vertical levels extending up to approximately 40 km altitude [Horowitz *et al.*, 2003]. MOZART-2 is built on the framework of the Model of Atmospheric Transport and Chemistry (MATCH). MATCH includes representations of advection, convective transport, boundary layer mixing, and wet and dry deposition. Penetrative convection in the NCEP Reanalysis 1 is described by Pan and Wu [1994], which is based on Arakawa and Schubert [1974] as simplified by Grell [1993] with a saturated downdraft. Shallow convection from NCEP Reanalysis 1 is determined by Tiedtke [1983]. We also forced MOZART-2 with meteorological data from the middle atmosphere version of NCAR Community Climate Model (MACCM3), which has the same convective scheme as the GEOS-4 Reanalysis. We found the CO₂ results from MOZART-2 forced by MACCM3 meteorological fields are very close to that from

GEOS-Chem driven by GEOS-4 data, so we defer the detailed discussion to a separate study.

[7] The GLOBALVIEW-CO₂ mixing ratio data [*Tans et al.* 1998; *GLOBALVIEW-CO₂*, 2002] are used in this study as the lower boundary condition for the Caltech/JPL CTM, GEOS-Chem, and MOZART-2. For convenience, we refer this hereforth as the GLOBALVIEW-CO₂ boundary condition. Since the GLOBALVIEW-CO₂ data are limited in space, especially over ocean, we used the GLOBALVIEW-CO₂ to rescale the CO₂ mixing ratio in the surface. First, we use seasonal varying CO₂ source and sink flux boundary condition to drive the model. We also interpolate monthly mean GLOBALVIEW-CO₂ measurements to GEOS-chem resolution, which are 2° in the latitude. Then, we rescale the zonal mean CO₂ mixing ratio in the boundary by the monthly mean GLOBALVIEW-CO₂ measurements for each month and for each latitudinal band. The monthly mean GLOBALVIEW-CO₂ flask data are close to the CO₂ GLOBALVIEW-CO₂ boundary condition when they are co-located. We assume that all atmospheric CO₂ originating from the surface layer is practically chemically inert in the atmosphere considering its long lifetime. With the GLOBALVIEW-CO₂ boundary condition, discrepancy between model results and observations would help diagnose potential issues with model transport. However, we noticed that the GLOBALVIEW-CO₂ surface stations are sparse in the southern hemisphere (SH), and that the GLOBALVIEW-CO₂ boundary condition is also biased toward oceanic sites.

[8] In a separate simulation using GEOS-Chem, we use prescribed CO₂ sources and sinks as the boundary condition, as described in *Suntharalingam et al.* [2004]. The exchange of CO₂ between the terrestrial biosphere and atmosphere is based on net primary productivity and respiration fluxes from the Carnegie-Ames-Stanford (CASA) ecosystem model [*Randerson et al.*, 1997]. Monthly mean biospheric CO₂ fluxes are used in the present study, for the inclusion of diurnal cycle appears to have relatively small effect on model CO₂ [*Suntharalingam et al.*, 2004]. In a sensitivity study, we include the diurnally varying biospheric CO₂ fluxes [*Olsen and Randerson*, 2004] in the GEOS-Chem from Feb 2000 to Dec 2000. The effects of the diurnal cycle in biospheric CO₂

fluxes on the upper tropospheric CO₂ sampled at Matsueda's aircraft locations are relatively small. This is because the aircraft data are taken over the ocean in the upper troposphere, which is away from the boundary layer. Air-to-sea exchange of CO₂ is from *Takahashi et al.* [1997]. Estimates of fossil fuel emissions are from *Marland et al.* [2007]. Monthly mean biomass burning emissions of CO₂ are derived based on *Duncan et al.* [2003]. The maximum contribution for the CO₂ seasonal cycle is from the exchange between the biosphere and atmosphere. Fossil fuel emission and biomass burning also have relatively large contribution to the CO₂ in the NH. Due to the upwelling in the ocean, ocean is a source for atmospheric CO₂ in the tropics. Ocean is a sink for CO₂ in the middle to high latitudes [*Takahashi et al.*, 1997]. Since there is an unbalanced CO₂ budget associated with the prescribed source and sink boundary condition [*Suntharalingam et al.*, 2003; *Suntharalingam et al.*, 2004], we constrain the restart file for the CO₂ mixing ratio in the beginning of each year by the GLOBALVIEW-CO₂ at the surface by regression. As a result, the unbalanced CO₂ budget is resolved in some degree. Discrepancies between the GEOS-Chem CO₂ simulations (driven by the same GEOS-4 reanalysis data) with the above-mentioned two boundary conditions would help identify potential issues with the surface sources and/or sinks on simulating CO₂ seasonal cycle.

[9] The details for the different model experiments are summarized in Table 1. Since Experiments A and B are forced by the same transport, GEOS-4 assimilated meteorology field, the only difference between the two experiments is the boundary conditions. Experiment A is forced by the GLOBALVIEW-CO₂ boundary condition, which is constrained by the monthly mean GLOBALVIEW-CO₂ surface flask data. Experiment B is forced by the flux boundary condition, in which we constrain the restart file for the CO₂ mixing ratio in January of each year by the GLOBALVIEW-CO₂ at the surface by regression method. Thus the main difference between the two experiments is the boundary condition from February to December in each year. Experiment A and Experiment E are forced by the same boundary conditions with different transport fields. The difference between Experiments A and E represents the difference in the transport fields. Model results will be compared with aircraft measurements from *Matsueda et al.* [2002] and GLOBALVIEW-CO₂ [*GLOBALVIEW-CO₂*, 2002] in Section 3. Aircraft CO₂

from *Matsueda et al.* [2002] are measured biweekly since April 1993 to present. The latitudinal coverage is approximately from 35°S to 35°N. The longitudinal coverage is from 135°E to 150°E. The CO₂ at 8-13 km over the western Pacific from Australia to Japan are measured. We also compared the model results with GLOBALVIEW-CO₂ aircraft measurements at Carr (40.9°N, 104.8°W) and Poker Flat (65.07°N, 147.29°W) in the auxiliary material.

3. Results

[10] To quantitatively compare the aforementioned aircraft observations and model results of CO₂, it is essential to separate trend from seasonal and semi-annual cycles in the data. For which a widely used approach is fitting data by a series of Legendre polynomials and harmonic functions [*Prinn et al.*, 2000]. We thus decompose CO₂ concentrations using the following empirical model:

$$X(t) = a + bNP_1(t/N - 1) + 1/3cN^2P_2(t/N - 1) + 1/5dN^3P_3(t/N - 1) + e\cos(2\pi t) + f\sin(2\pi t) + g\cos(4\pi t) + h\sin(4\pi t) \quad (1)$$

where t is from 0 to the $2N$ year (whole time period); P_1 , P_2 , and P_3 are the first, second, and third Legendre polynomials. The coefficients a , b , c , and d are the mean value, the trend, the acceleration in the trend, and the coefficient for P_3 , respectively. We add the third Legendre function to better fit the data sets. The harmonic functions are added for seasonal and semi-annual cycles. e and f are the amplitudes of the annual cycle, while g and h are the amplitudes of the semi-annual cycle. Seasonal cycle amplitude ($\sqrt{e^2 + f^2}$) for CO₂ is listed in Table 2.

[11] Figure 1 compare the aircraft observations of CO₂ averaged between 8 and 13 km (red dots) [*Matsueda et al.*, 2002] and model results averaged at the same altitude range for 2000-2004. The panels are for 35°S to 35°N latitudes in 10° steps. CO₂ from all 3-D CTMs are sampled as the same location as the aircraft data. The zonal mean CO₂ from the 2-D CTM are compared with the aircraft data directly. The amplitudes of the seasonal cycle of CO₂ are smaller in the SH than those in the NH, for there is less contribution from the seasonal cycle in the vegetation photosynthesis. The green dashed line shows

results from a GEOS-Chem simulation driven by GEOS-4 data using the GLOBALVIEW-CO₂ boundary condition (Experiment A in Table 1). The orange dash-dotted line is GEOS-Chem CO₂ (driven by GEOS-4 data) with prescribed sources and sinks (Experiment B). Results from these two simulations are generally consistent, except that the CO₂ seasonal cycle is smaller in Experiment B than that in Experiment A (See Table 2). Because the transport is the same in both experiments, difference in the results may reflect deficiencies in the prescribed sources/sinks in the summer in Experiment B. In general, CO₂ concentrations from Experiment B are larger than those from Experiment A in the NH from July to October, especially at 35°N. This is consistent with a possible missing terrestrial sink hypothesis in the NH by *Tans et al.* [1990]. The GEOS-Chem CO₂ forced by the GEOS-3 meteorological fields and the GLOBALVIEW-CO₂ boundary condition (pink dotted line; Experiment C) and CO₂ from MOZART-2 (blue long dash-dotted line; Experiment E) both agrees reasonably well with the aircraft data, except some underestimations of CO₂ seasonal cycle in the NH. Experiment C includes only results for 2000-2002, as GEOS-3 data are available for only up to 2002.

[12] The agreement between the 3-D model results and aircraft data is fairly good, except at the NH mid-latitudes, where the 3-D models underestimate the amplitude of the seasonal cycle of CO₂ as seen in the aircraft data, which are consistent with results found in column-averaged CO₂ by *Yang et al.* [2007]. Similar results are found by comparing the model results to the GLOBALVIEW-CO₂ aircraft CO₂ data at Carr (40.9°N, 104.8°W) and Poker Flat (65.07°N, 147.29°W) as shown in the Figs. A1-A2 of the auxiliary material. In fact, the models all tend to underestimate the seasonal cycles of CO₂ in the middle to high latitudes.

[13] To investigate this problem, we plotted the vertical profiles of CO₂ simulated by each model at 5°N (upper panel) and 35°N (lower panel) of 2003 in Figure 2. For a fair comparison between 2-D and 3-D models, we calculate the zonal mean CO₂ from all 3-D models. In the tropics (5°N) the 3-D model results closely follow that from the 2-D model. The main advantage of the 2-D model over the 3-D models appears to be a stable numerical scheme and the flexibility to fine-tune the transport in the model. The 2-D

model has been tuned to reproduce the age of air to within the errors of the measurements in the stratosphere [see, *e.g.*, Appendix A of *Morgan et al.*, 2004] and to simulate CO₂ reasonably well in the upper troposphere [*Shia et al.*, 2006]. In the northern mid-latitudes (35°N), all 3D models seem to underestimate the upper tropospheric CO₂ in January and April of 2003. In the later part of the paper, we will discuss the key parameter that affects the upper tropospheric CO₂ most. CO₂ simulations from MOZART-2 (Experiment E) and GEOS-Chem with GLOBALVIEW-CO₂ boundary condition (Experiment A) are forced by the same boundary condition. Thus the difference of CO₂ vertical profiles produced by Experiments A and E shows that the transport is very important for CO₂ in mid-troposphere.

[14] Raw CO₂ data from the aircraft measurements and model experiments at 35°N are shown as red dots and solid lines respectively in Figure 3. Dashed lines are the sum of all terms in the right hand side of Eq. (1), which fit well with the raw aircraft data and model results. We then detrended the data by subtracting the sum of the first three Legendre functions. The results are very close to remove a third order polynomials. The detrended aircraft data in the four years are shown as red dots in Figs. 4 and 5. Diamond and error bar are the mean and standard deviation of the detrended aircraft data for each month. Black dotted line is the sum of the annual and semi-annual cycles terms in Eq. (1), which follows well the monthly mean aircraft data (Diamonds). For comparison, we also detrended the model results using the same method. Then we averaged the detrended model CO₂ from all four years. Results are shown as color lines in Figs. 4 and 5. The phase of CO₂ seasonal cycle is well captured by the different model simulations. The seasonal cycle amplitude is larger in the NH than that in the SH, which is captured by all models. Most 3-D models underestimate the seasonal cycle amplitude in the NH.

[15] The latitudinal distribution of CO₂ seasonal cycle amplitude is shown in Fig. 6. Because of the short simulation time period in Experiment C, we do not include it in Fig. 7. All 3-D models underestimate the amplitude of CO₂ seasonal cycle in the NH mid-latitudes. The seasonal cycle amplitude of upper tropospheric CO₂ in the 2-D CTM is larger than those from the 3-D models. The amplitude of CO₂ seasonal cycle is larger in

MOZART-2 than those in GEOS-Chem. The GEOS-Chem simulation forced by the GLOBALVIEW-CO₂ boundary condition (Experiment A) has a larger CO₂ seasonal cycle than the GEOS-Chem simulation forced by surface sources and sinks (Experiment B). In Figure 5, the CO₂ at 35°N from Experiment B (orange line) are larger than that from Experiment A (green line) during July to October. This indicates that the biospheric CO₂ flux in the NH might be too strong in Experiment B. The result is consistent with possible missing terrestrial sinks in the NH suggested by *Tans et al.* [1990]. GEOS-Chem CO₂ (Experiments A and B) overestimate the seasonal cycle amplitude in the SH, which may be due to biases in the SH transport in GEOS-4 [*Bloom et al.*, 2005]. Since there are fewer rawinsonde data in the SH compared with those in the NH, the transport in the SH is less constrained in the GEOS-4 assimilated data [*Bloom et al.*, 2005].

[16] To further explore the role of different parameters for simulating CO₂ correctly in the upper troposphere, sensitivity studies have been conducted using the GEOS-Chem model driven by GEOS-4 reanalysis data and the GLOBALVIEW-CO₂ boundary condition. We first perturbed the turbulent mixing in the planetary boundary layer by 50% through increasing the turbulent mixing coefficient (Experiment F). The resulting differences between the perturbed run (Experiment F) and control run (Experiment A) at 35°N are shown in Fig. 7a. The CO₂ concentrations differ by less than ~0.04 ppmv at altitudes below 3.5 km, a rather small effect. We also perturbed separately the convective updraft mass flux by 20% (Experiment G). The resulting differences between the perturbed run (Experiment G) and control run (Experiment A) at 35°N are shown in Fig. 7b. The largest increase of ~0.65 ppmv in CO₂ is found at 6 km, which is very significant for simulating the upper tropospheric CO₂. Compared with the turbulent mixing in the boundary layer, the convective mass flux is more important for lifting the CO₂ from surfaces to the middle and upper troposphere.

[17] Accurate simulation of CO₂ concentrations in the upper troposphere is also imperative for deducing the inter-hemispheric transport of CO₂. It is generally accepted that the NH is a net CO₂ source and the SH (the oceans) is a net CO₂ sink [*IPCC*, 2001]. Previous studies [*Prather et al.* 1987; *Prinn et al.*, 1992] and recent study on

interhemispheric transport of GEOS-Chem CO [Cai *et al.*, 2008] suggest that a large component of the inter-hemispheric transport occurs in the upper troposphere. Therefore, correctly modeling upper tropospheric CO₂ takes on added significance. Consider a flux inversion in which CO₂ in the NH was not efficiently transported to the upper troposphere, resulting in less transport to the SH and a lower calculated southern ocean sink. This would create artificially high CO₂ in the NH, demanding a large land sink to reconcile the model predictions with the observations. Such arguments have recently been advanced by Stephens *et al.* [2007] and Baker [2007].

4. Conclusions

[18] 2-D and 3-D chemistry and transport models, driven by different transport schemes, have been used to simulate the upper tropospheric CO₂ from 2000 to 2004. We also apply different boundary conditions to force the 3-D CTMs. We found that the transport schemes are very important for simulating the upper tropospheric CO₂. Model CO₂ agree generally well with the aircraft data from 35°S to 35°N. The trends of CO₂ are simulated correctly by most of the models. The phases of CO₂ seasonal cycles are also captured well by models. Similar to those in the aircraft data, model CO₂ have a smaller seasonal cycle amplitudes in the SH compared with those in the NH. However, 3-D CTMs appear to underestimate the seasonal cycle amplitude of upper tropospheric CO₂ in the NH mid-latitudes. Sensitivity studies reveal that the convective mass fluxes are very crucial for simulating the upper tropospheric CO₂. In addition to the aircraft data, global AIRS CO₂ data will become available in the near future [Chahine *et al.*, 2008]; global total column CO₂ data will be available in two years [Crisp *et al.*, 2004]. These data can be used to constrain the vertical and horizontal transport in the CTMs, resulting in more realistic models. This will give us greater confidence in deducing sources and sinks of CO₂ using a combination of global CO₂ data and inverse modeling [Miller *et al.*, 2007].

Acknowledgement: This work is performed at JPL under contract with NASA, and is supported by project 102438-01.01.01. We thank J. Randerson for useful inputs and

helpful comments, two anonymous reviewers and C. L. Quere for helpful comments. Our special thanks to Bob Yantosca at Harvard University for the help on the GEOS-Chem model and to Peter Hess, Larry Horowitz, and Jean-Francois Lamar on the MOZART-2 model. R. L. Shia and Y. L. Yung are supported by NSF grant ATM-9903790. M.C. Liang also would like to acknowledge the support from an NSC grant 96-2628-M-001-018 to Academia Sinica.

References:

- Allen, D. J., R. B. Rood, A. M. Thompson, and R. D. Hidson (1996a), Three-dimensional ^{222}Rn calculations using assimilated data and a convective mixing algorithm, *J. Geophys. Res.*, *101*, 6871-6881.
- Allen, D. J., *et al.* (1996b), Transport induced interannual variability of carbon monoxide using a chemistry and transport model, *J. Geophys. Res.*, *101*, 28655-28670.
- Anderson, B. E. *et al.* (1996), Airborne observations of spatial and temporal variability of tropospheric carbon dioxide, *J. Geophys. Res.*, *101*(D1), 1985-1997.
- Andres, R. J., G. Marland, I. Fung, and E. Matthews (1996), Distribution of carbon dioxide emissions from fossil fuel consumption and cement manufacture, 1950-1990, *Global Biogeochem. Cycles*, *10*, 419-429.
- Arakawa, A., and W. H. Shubert (1974), Interaction of a cumulus ensemble with the large-scale environment, Part I, *J. Atmos., Sci.*, *31*, 674-704.
- Baker, D. F. (2007), Reassessing carbon sinks, *Science*, *316*, 1708-1709.
- Bloom, S., *et al.* (2005), The Goddard Earth Observation System Data Assimilation System, GEOS DAS Version 4.0.3: Documentation and Validation, NASA TM-2005-104606, 26, 166pp.
- Cai, C., *et al.* (2008), A satellite perspective on the interhemispheric transport of pollution, *In Preparation*.
- Chahine, M., *et al.*, (2005), On the determination of atmospheric minor gases by the method of vanishing partial derivatives with application to CO_2 , *Geophys. Res. Lett.*, *32*(22), L22803.
- Chahine, M., *et al.*, (2008), First satellite remote sounding of the global distribution of mid-tropospheric CO_2 , *In Preparation*.
- Crisp, D., *et al.*, (2004), The orbiting carbon observatory (OCO) mission, *Advances in Space Research*, *34*, 700-709.
- Dickinson, R. E., and R. J. Cicerone (1986), Future global warming from atmospheric trace gases, *Nature*, *319*(6049), 109-115.
- Duncan, B. N., *et al.* (2003), Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, *J. Geophys. Res.*, *108*, doi:10.1029/2002JD002378.
- Fan, S., *et al.* (1998), A large terrestrial carbon sink in North America implied by atmospheric and oceanic CO_2 data and models, *Science*, *282*, 442-446.
- Fleming, E. L., *et al.* (2002), Two-dimensional model simulations of the QBO in ozone and tracers in the tropical stratosphere, *J. Geophys. Res.*, *107*, doi:10.1029/2001JD001146.
- GLOBALVIEW- CO_2 (2007), *Cooperative Atmospheric Data Integration Project: Carbon Dioxide* [CD-ROM], NOAA ESRL, Boulder, Colorado (Also available on Internet via anonymous FTP to <ftp.cmdl.noaa.gov>, Path: ccg/co2/GLOBALVIEW)
- Gloor, M. (2000), Optimal sampling of the atmosphere for purpose of inverse modeling: A model study, *Global Biogeochem. Cycles*, *14*(1), 407-428.
- Grell, G. A. (1993), Prognostic evaluation of assumptions used by cumulus parameterizations, *Mon. Wea. Rev.*, *121*, 764-787.

- Gurney, K. R., *et al.* (2003), TransCom 3 CO₂ inversion intercomparison: 1. Annual mean control results and sensitivity to transport and prior flux information, *Tellus, Ser. B*, 55, 555-579.
- Gurney, K. R. *et al.* (2004), Transcom 3 inversion intercomparison: Model mean results for the estimation of seasonal carbon sources and sinks, *Global Biogeochem. Cycles*, 18(1), GB1010.
- Kalnay, E., *et al.* (1996), The NCEP/NCAR 40-year reanalysis project, *Bull. Am. Meteorol. Soc.*, 77, 437-471.
- Kawa, S. R., *et al.* (2004), Global CO₂ transport simulations using meteorological data from the NASA data assimilation system, *J. Geophys. Res.*, 109, doi:10.1029/2004JD004554.
- Hack, J. J. (1994), Parameterization of moist convection in the NCAR community climate model (CCM2), *J. Geophys. Res.*, 99, 5551-5568.
- Holtzlag, A., and B. Boville (1993), Local versus nonlocal boundary-layer diffusion in a global climate model, *J. Clim.*, 6, 1825-1842.
- Horowitz, L. W., *et al.* (2003), A global simulation of tropospheric ozone and related tracers: description and evaluation of MOZART, version 2, *J. Geophys. Res.*, 108, doi:10.1029/2002JD002853.
- IPCC (2001), *IPCC Third Assessment Report (TAR): Climate Change 2001*, Intergovernmental Panel on Climate Change.
- Jiang, X. C. D. Camp, R. Shia, D. Noone, C. Walker, and Y. L. Yung (2004), Quasi-biennial oscillation and quasi-biennial oscillation-annual beat in the tropical total column ozone: A two-dimensional model simulation, *J. Geophys. Res.*, 109, Art. No. JD004377.
- Law, R., *et al.* (1996), Variations in modeled atmospheric transport of carbon dioxide and the consequences for CO₂ inversions, *Global Biogeochem. Cycles*, 10, 783-796.
- Lin, S. J., and R. B. Rood (1996), Multidimensional flux form semi-Lagrangian transport schemes, *Mon. Weather Rev.*, 124, 2046-2070.
- Marland, G., T. A. Boden, and R. J. Andres, Global, Regional, and National CO₂ Emissions. In *Trends: A Compendium of Data on Global Change. Carbon Dioxide Information Analysis Center*, Oak Ridge National Laboratory, U.S. Department of Energy, (Oak Ridge, TN, 2007).
- Matsueda, H., H. Y. Inoue, and M. Ishii (2002), Aircraft observation of carbon dioxide at 8-13 km altitude over the western Pacific from 1993 to 1999, *Tellus*, 54B(1), 1-21. The data are available at <http://gaw.kishou.go.jp/wdcgg.html>
- Miller, C. E., *et al.* (2007), Precision requirements for space-based Xco₂ data, *J. Geophys. Res.*, 112, doi:10.1029/2006JD007659.
- Moorthi, S., and M. J. Suarez (1992), Relaxed Arakawa-Schubert – A parameterization of moist convection for general circulation models, *Mon. Wea. Rev.*, 120(6), 978-1002.
- Morgan, C. G., *et al.* (2004), Isotopic fractionation of nitrous oxide in the stratosphere: Comparison between model and observations, *J. Geophys. Res.*, 109, D04305.
- Nakazawa, T., *et al.* (1997), Aircraft measurements of the concentrations of CO₂, CH₄, N₂O, and CO and the carbon and oxygen isotopic ratios of CO₂ in the troposphere over Russia, *J. Geophys. Res.*, 102, 3843-3859.

- Olsen, S. C., and J. T. Randerson (2004), Differences between surface and column atmospheric CO₂ and implications for carbon cycle research, *J. Geophys. Res.*, *109*, doi:10.1029/2003JD003968.
- Pan, H. L., and W. S. Wu (1994), Implementing a mass flux convection parameterization package for the NMC medium-range forecast model, *NMC office note*, No. 409, 40pp.
- Prather, M., *et al.* (1987), Chemistry of the global troposphere: Fluorocarbons as tracers of air motion, *J. Geophys. Res.*, *92*, 6579-6613.
- Prinn, R. G., *et al.* (1992), Global average concentration and trend for hydroxyl radicals deduced from ALE/GAGE trichloroethane (methylchloroform) data for 1978-1990, *J. Geophys. Res.*, *97*, 2445-2461.
- Prinn, R. G., *et al.* (2000), A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, *J. Geophys. Res.*, *105*, 17751-17792.
- Randerson, J. T., *et al.* (1997), The contribution of terrestrial sources and sinks to trends in the seasonal cycle of atmospheric carbon dioxide, *Global Biogeochem. Cycles*, *11*, 535-560.
- Shia, R., M. Liang, C. E. Miller, and Y. L. Yung (2006), CO₂ in the upper troposphere: influence of stratosphere-troposphere exchange, *Geophys. Res. Lett.*, *33*, doi:10.1029/2006GL026141.
- Stephens, B. B., *et al.* (2007), Weak northern and strong tropical land carbon uptake from vertical profiles of atmospheric CO₂, *Science*, *316*, 1732-1735.
- Suntharalingam, P., *et al.* (2003), Estimating the distribution of terrestrial CO₂ sources and sinks from atmospheric measurements: sensitivity to configuration of the observation network, *J. Geophys. Res.*, *108*, doi:10.1029/2002JD002207.
- Suntharalingam, P., *et al.* (2004), Improved quantification of Chinese carbon fluxes using CO₂/CO correlations in Asian outflow, *J. Geophys. Res.*, *109*(D18), doi:10.1029/2003JD004362.
- Takahashi, T., *et al.* (1997), Global air-sea flux of CO₂: An estimate based on measurements of sea-air pCO₂ difference, *Proc. Natl. Acad. Sci. U. S. A.*, *94*, 8929.
- Tans, P. P., I. Y. Fung, and T. Takahashi (1990), Observational constraints on the global atmospheric CO₂ budget, *Science*, *247* (4949): 1431-1438.
- Tans, P.P., *et al.* (Eds.) (1998), Carbon Cycle, in *Climate Monitoring and Diagnostics Laboratory No. 24 Summary Report 1996-1997*, edited by D.J. Hoffmann *et al.*, chap. 2, pp. 30-51, NOAA Environ. Res. Lab., Boulder Colo. (Data available from <http://www.cmdl.noaa.gov/>)
- Tiedtke, M. (1983), The sensitivity of the time-mean large-scale flow to cumulus convection in the ECMWF model, *ECMWF workshop on convection in large-scale models*, Reading, England, pp. 297-316.
- Yang, Z. *et al.* (2007), New constraints on northern hemisphere growing season net flux, *Geophys. Res. Lett.*, *34*, doi:10.1029/2007GL029742.
- Zhang, G. J., and N. A. McFarlane (1995), Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian climate centre general circulation model, *Atmos. Ocean*, *33*, 407-446.

Figure Captions:

Figure 1: Aircraft observations between 8 km and 13 km (red dots) [Matsueda *et al.*, 2002] and modeled CO₂ mixing ratios averaged at the same layer from 2000 to 2004. The panels are for 35°S, 25°S, 15°S, 5°S, 5°N, 15°N, 25°N, and 35°N, respectively. The CO₂ mixing ratios from the GEOS-Chem model (Experiments A, B, and C) are shown by the green dashed, orange dash-dotted, and pink dotted lines, respectively. The CO₂ mixing ratios from the Caltech-JPL 2-D model (Experiment D) are shown by purple solid lines. The CO₂ mixing ratios from MOZART-2 (Experiment E) are shown by the blue long dash-dotted lines.

Figure 2: Vertical profiles of CO₂ in January, April, July, and October 2003. Colors are the same as in Figure 1. Upper panel: Latitude = 5°N. Lower panel: Latitude = 35°N.

Figure 3: CO₂ from aircraft and models at 35°N. Red dots are aircraft observations. Solid lines are model results. Colors are the same as in Figure 1. Dashed lines are the fit to the CO₂ (see text).

Figure 4: CO₂ seasonal cycles from detrended aircraft (Red dots) and detrended model results (Color lines) at 25°S, 15°S, and 5°S. Trends are determined by the sum of the first three legendre polynomials. Diamond and the error bar are the mean and standard deviation of the detrended aircraft data within each month. Black dotted line is the sum of the annual and semi-annual cycles terms in Eq. (1).

Figure 5: CO₂ seasonal cycles from detrended aircraft (Red dots) and detrended model results (Color lines) at 5°N, 15°N, 25°N, and 35°N. Trends are determined by the sum of the first three legendre polynomials. Diamond and error bar are the mean and standard deviation of the detrended aircraft data within each month. Black dotted line is the sum of the annual and semi-annual cycles terms in Eq. (1).

Figure 6: Latitudinal distribution of CO₂ seasonal cycle amplitude.

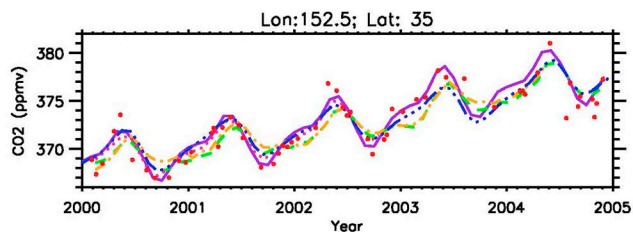
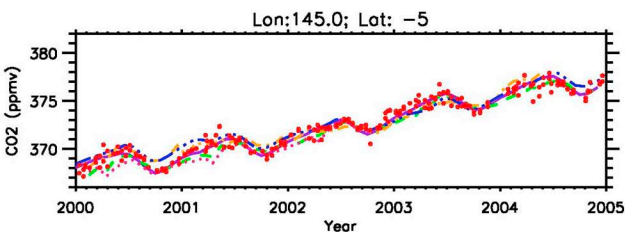
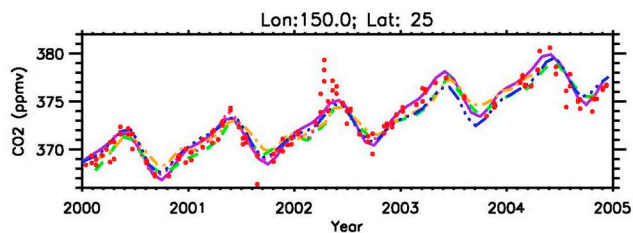
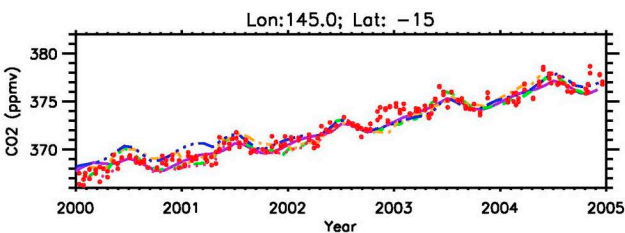
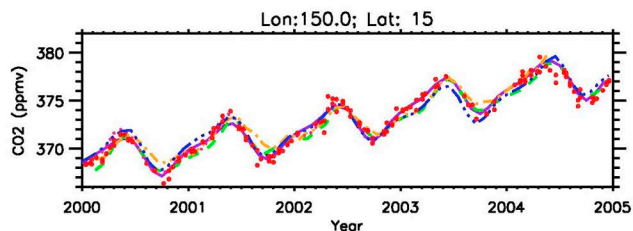
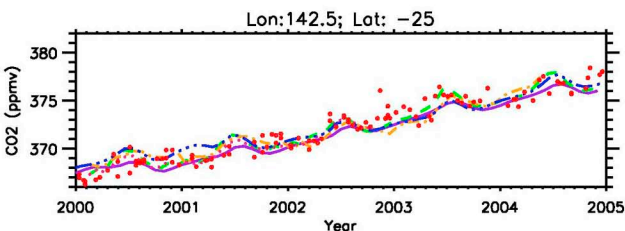
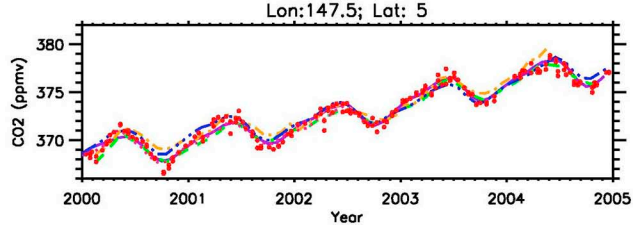
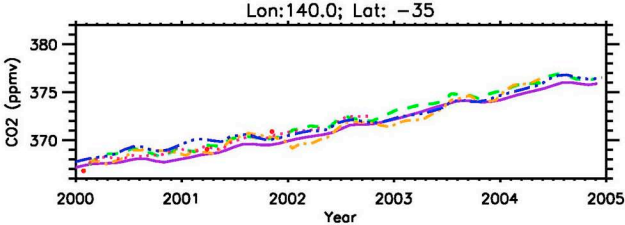
Figure 7: (a) CO₂ difference between the enhanced turbulence mixing in the planetary boundary layer simulation and control experiment at 35°N. (b) CO₂ difference between the enhanced convective updraft mass flux simulation and control experiment at 35°N.

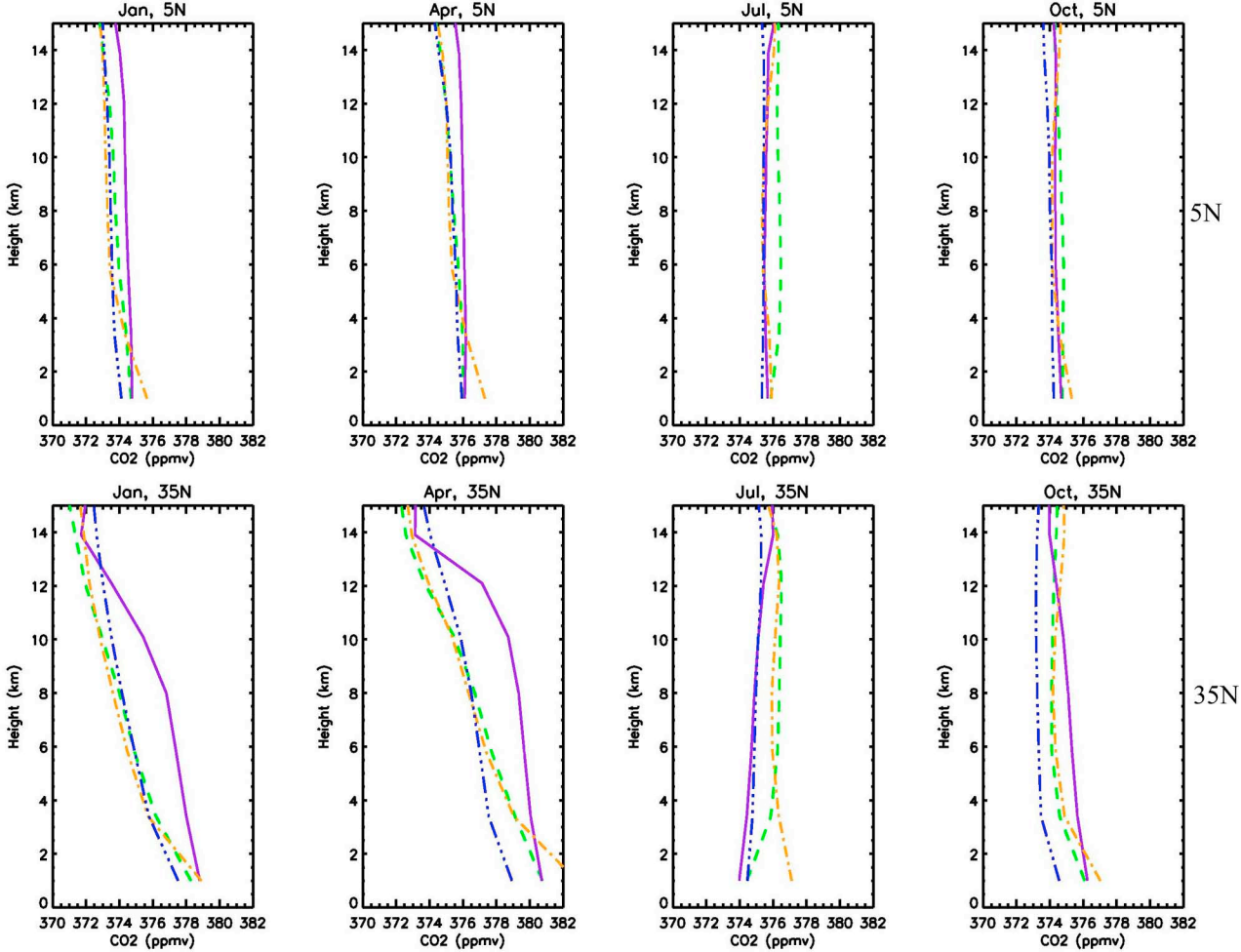
Table 1: Description of Model Experiments.

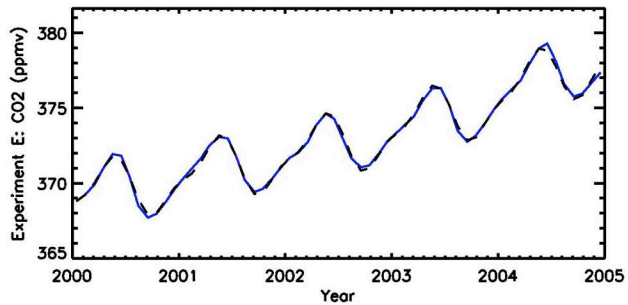
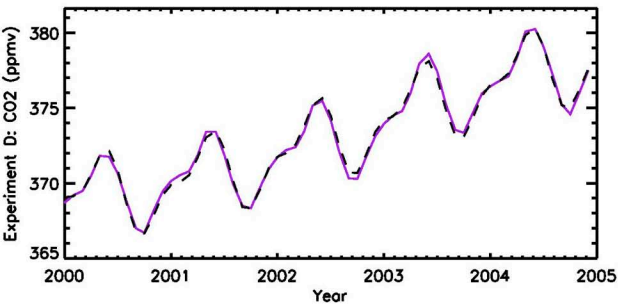
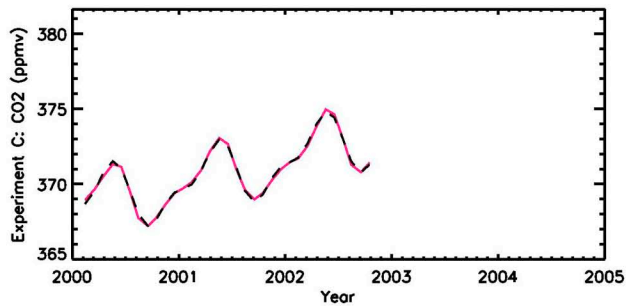
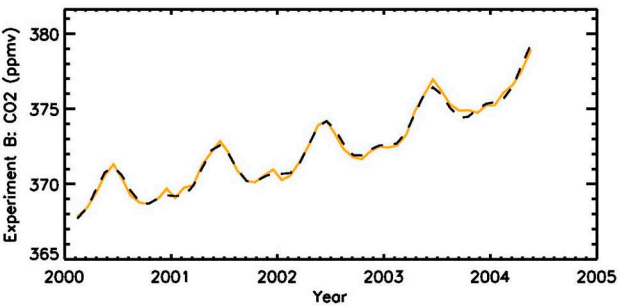
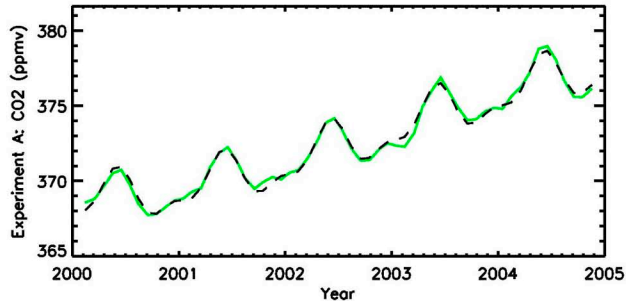
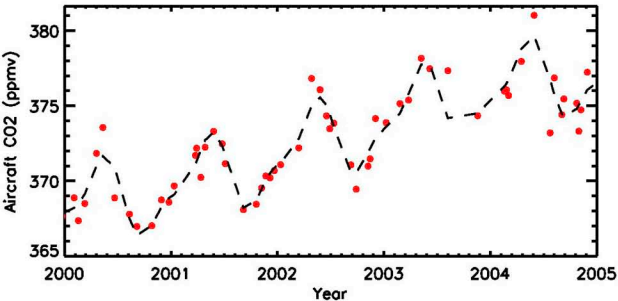
	Model	Transport	Boundary Condition	Model Change
Experiment A	3-D GEOS-Chem	GEOS-4	GLOBALVIEW-CO ₂	
Experiment B	3-D GEOS-Chem	GEOS-4	CO ₂ sources and sinks	
Experiment C	3-D GEOS-Chem	GEOS-3	GLOBALVIEW-CO ₂	
Experiment D	2-D Caltech/JPL CTM	NCEP2 and UKMO	GLOBALVIEW-CO ₂	
Experiment E	3-D MOZART2	NCEP1	GLOBALVIEW-CO ₂	
Experiment F	3-D GEOS-Chem	GEOS-4	GLOBALVIEW-CO ₂	Increase turbulence mixing in the PBL by 50%
Experiment G	3-D GEOS-Chem	GEOS-4	GLOBALVIEW-CO ₂	Increase the convective updraft mass flux by 20%

Table 2: Seasonal Cycle Amplitude of CO₂ From Matsueda Aircraft data and Model Simulations.

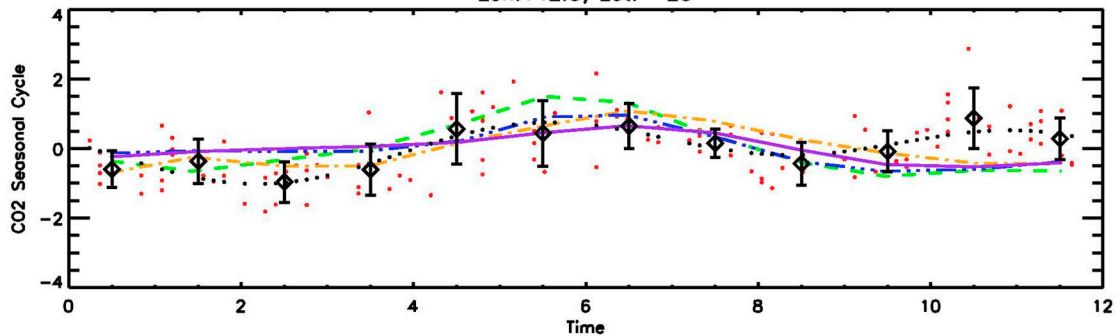
	Seasonal Cycle Amplitude (ppmv)							
Aircraft		0.38±0.14	0.38±0.08	0.90±0.08	1.61±0.08	2.05±0.09	2.47±0.17	2.48±0.3
Experiment A	0.28±0.06	0.95±0.08	1.03±0.1	0.74±0.08	1.21±0.07	1.64±0.06	1.61±0.06	1.51±0.07
Experiment B	0.57±0.08	0.7±0.09	0.74±0.1	0.57±0.1	1.17±0.07	1.5±0.07	1.3±0.06	1.32±0.07
Experiment C	0.1±0.08	0.64±0.07	0.83±0.1	0.52±0.1	1.24±0.11	1.98±0.09	2.06±0.06	1.87±0.05
Experiment D	0.21±0.05	0.49±0.07	0.66±0.08	1.06±0.07	1.44±0.07	1.94±0.07	2.33±0.07	2.36±0.08
Experiment E	0.28±0.05	0.58±0.05	0.64±0.06	0.77±0.06	1.30±0.05	1.88±0.06	1.90±0.07	1.88±0.06



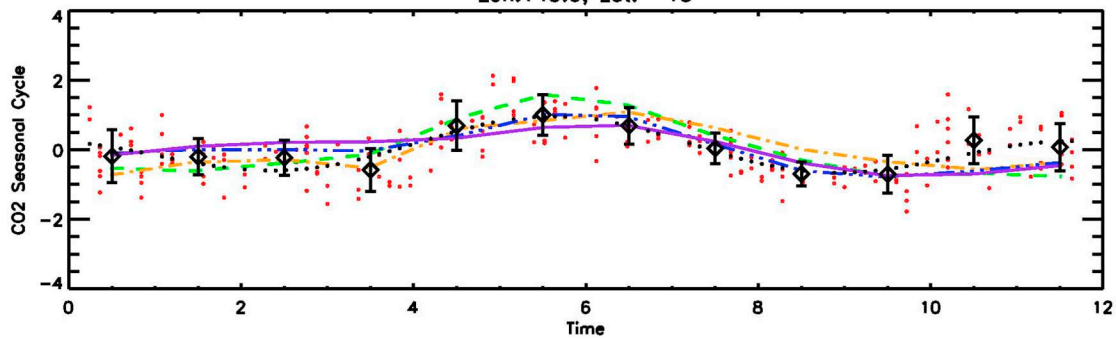




Lon:142.5; Lat: -25



Lon:145.0; Lat: -15



Lon:145.0; Lat: -5

